BOND COAT CONSIDERATIONS FOR THERMAL BARRIER COATINGS

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Abstract

Bond coat oxidation has been clearly established as a factor contributing to the failure of a TBC. However, the mechanism by which oxidation effects TBC failure has not been determined. Similarly, other bond coat properties have been cited as influencing TBC residual stresses, but neither the magnitude of the effects nor the exact mechanisms by which these properties act, and interact, have been elucidated. A finite element model was developed to evaluate residual stresses induced by thermal cycling of a typical plasma sprayed TBC system. It was found that bond coat properties of oxidation, creep, coefficient of thermal expansion (CTE) and roughness, as well as top coat creep, all strongly influence ceramic layer stresses. Roughness and CTE were found to have the strong effects in a parametric study, acting primarily as stress multipliers for other active mechanisms. A detailed examination of the time dependent properties of creep and oxidation indicates that these factors account for evolution of the residual stresses with thermal cycling. It is the combination of these effects over the lifetime of the coating that drives the generation of stresses required to initiate and propagate delamination cracks.

Introduction

It is universally acknowledged that bond coats for plasma sprayed TBCs have a strong effect on the thermal fatigue life of the ceramic layer, but the specific mechanisms by which the bond coat influences TBC life have not been established [1, 2, 3, 4]. As an example, bond coat oxidation has been clearly linked to spallation of the ceramic top coat but the mechanism by which oxidation causes failure of the top coat has not been clearly identified [1, 4, 5]. Similarly, bond coat coefficient of thermal expansion (CTE), roughness and creep properties have been proposed to influence TBC failure, but the magnitude of the effects and the physical mechanisms by which these effects act, and interact, have not been established [1, 2, 6].

Part of the difficulty in determining which mechanisms and interactions are involved is due to the difficulty in isolating variables in a real material system so that unambiguous results can be obtained. An alternative is to perform modeling studies to determine trends in the effects of various factors as well as determine the mechanisms that precipitate failure. There have been only a few models that have focused in this important area for TBCs. These models have examined the TBC residual stresses resulting from single cooling cycles from a stress free state [6, 7] and residual stresses during thermal cycling [8]. but have all been incomplete in that some known factors were not included. The purpose of the current work was to develop a more inclusive finite element model to characterize the effects of TBC system properties, and particularly bond coat properties, on the residual stresses in TBCs. This was accomplished by adding cyclic oxidation and multiple thermal cycle capability to the previous Petrus model [8], which had already incorporated the effects of creep, CTE and roughness on TBC stresses during a single thermal cycle. As will be shown, the results of this inclusive model indicate that creep processes dominate the generation of ceramic layer stress early in the life of a TBC, oxidation dominates stress generation later in life, but that there is interaction between all the effects throughout coating life that cannot be ignored.

2. Methodology

This study used NIKE 2-D stress models [9] to describe a burner rig test of a 2.54 cm (1.0 in.) diameter solid Waspaloy rod coated with 0.013 cm (0.005 in.) of NiCrAlY bond coat, which was in turn coated with a "top coat" of 0.025 cm (0.010 in.) ZrO_2-8wt.% Y_2O_3 (Figure 1(a)). It was assumed that the coating was stress free after deposition and cooling to room temperature a reasonable assumption in view of the low in-plane compressive stresses (approximately 3 MPa) measured on TBCs fabricated with appropriate temperature control [10]. The model geometry was a thin axisymmetric slice of the substrate coating system, as was used previously (Figure 1(a)) [8], with a condition of generalized plane strain. Unlike the Petrus model [8], the current model used refined elements for the bond coat and top coat at the interface region to allow for growth and analysis of an oxide layer. The

geometry of the bond coat/ceramic interface was modeled as a sine wave to simulate circumferential surface roughness conditions typical for a plasma sprayed TBC (Figure 1(b).) [6, 7, 8].

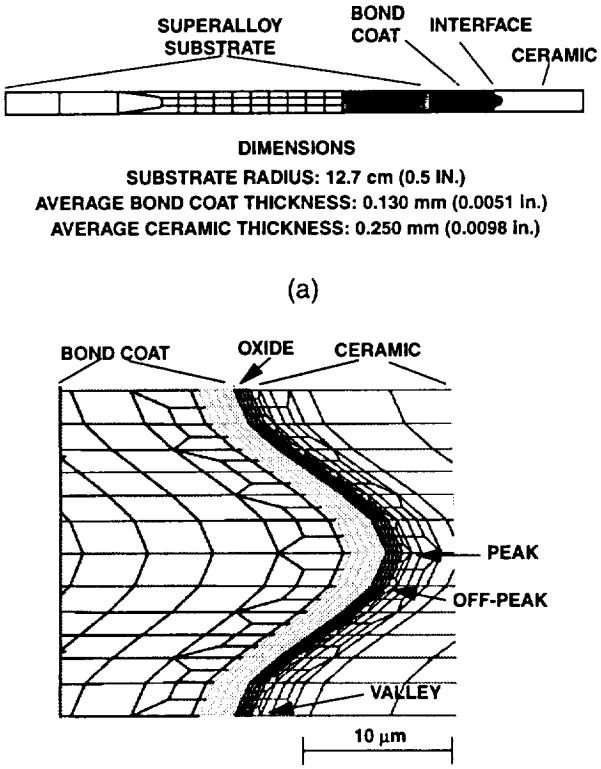


Figure 1. Finite element mesh geometry for TBC oxidation model showing (a) overall geometry of axisymmetric slice and (b) detail view of the interface region.

The thermal cycle profile for the model was based on the actual burner rig profile (Figure 2) [8]. The model included the temperature gradients that naturally occur during heating and cooling. It should be noted that the temperature gradient was approximately zero at the hold temperature since the modeled specimens were not internally cooled.

Creep behavior was defined using a temperature dependent power-law creep model, with creep behavior as defined in the literature [2, 11, 12]. Further properties information required for the model included the temperature dependent properties of bulk modulus, shear modulus and coefficient of thermal expansion, and were derived from the literature [2, 11, 12, 13].

The two levels for oxidation were defined as a low of no oxidation and a high equal to the oxidation rate found empirically for the alumina former NiCoCrAlY [11]. The oxide scale was assumed to be of uniform

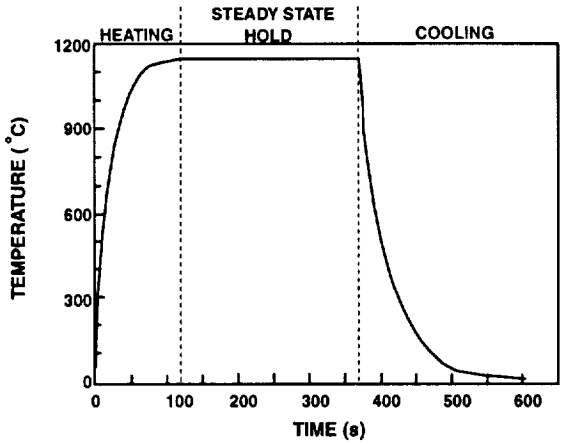


Figure 2. Measured burner rig test thermal profile used in the FEM model.

thickness across the entire interface and was further assumed to be composed entirely of alumina. The properties used for the CTE and modulus of the scale were those for a sintered 95% dense alumina [13]. The scale was assumed to behave elastically under all conditions.

The procedure for incorporating a growing oxide on the bond coat used the element "birth" and "death" capabilities of NIKE. Bond coat oxidation was thus modeled through the replacement of bond coat elements with the same size of oxide elements at set time intervals. The first oxide birth was in the elements located at the ceramic metal interface. Subsequent growth occurred through oxide elements birthed at the scale/ metal interface at the start of the high temperature hold portion of the cycle. The oxide elements were allowed to "grow" during steady state to the full volume dictated by the growth rate equation by assigning an artificially high CTE to the oxide and imposing a small temperature increase during steady state, similar to the method used by Chang et al. [6]. In this way the stress due to oxide growth, relaxation of growth stresses and thermal cycling was incorporated in the model. Since the oxide elements were birthed at the oxide/bond coat interface, the model simulates the inward growth of an oxide scale.

Five factors were intentionally varied, in addition to the main factor of oxidation. The five factors were bond coat roughness, bond coat CTE, bond coat creep, top coat creep and the number of thermal cycles. Information on stress as a function of location is also a part of the output of the model. After a few brief

comments on the effects of roughness and CTE, the remainder of the paper will concentrate on the effects of oxidation and creep of the constituents.

The output of the model included stress as a function of time, location and direction. It was found that the radial stresses in the ceramic (perpendicular to the interface) provided an adequate indication of the stresses most likely to promote delamination cracking. Therefore, radial stresses will be presented as a function of time and location to depict the active stress generating mechanisms in a TBC system and to try to define the likely failure scenario. It was also found (see Figures 8 and 9, discussed later) that the highest tensile radial stresses were located at the bond coat peak, off-peak and valley regions. Therefore, the large volume of data generated during FEM modeling will be narrowed to these locations in order to facilitate comparison of the various cases presented.

3. Results and Discussion

The first point to note in the discussion is that the intent of the model at this stage of development is to examine mechanisms that generate residual stresses (or strains) in TBCs. A reasonable assumption is that high residual stresses are a cause of ceramic failure. Thus, knowing how the residual stresses are generated provides insight on how the coating fails. The intent of a "final" model is to relate the stress generating mechanisms to failure of the TBC. Thus, the important points to follow in this paper are the trends in residual stress development with the selected factors, not the absolute values of the stresses. With this in mind, it is informative to compare the calculated stresses to those measured in real systems. Coating stress measurements are of the stress in the plane of the coating, not the stresses normal to the plane. For zirconia, these stresses are quite low for the as-deposited coating, between 0 and 5 MPa. After one cycle, the in-plane stresses typically jump to 20 to 30 MPa, and jump to 25 to 35 MPa after 10 one hour cycles to 1100°C [10]. In the current study, the average compressive residual stresses in the plane of the coating have been calculated to be as high as 200 MPa after 4 cycles if coating cracking is not allowed, in obvious disagreement with the measured values. However, when the coating is allowed to crack in the model, the average coating stresses after 4 cycles drop to approximately 20 MPa. This comparison indicates: the calculated stress is sensitive to the fracture stress of the coating, the model is capable of predicting realistic stresses when fracture is included, it is best to use the stresses reported in the current paper as an indication of the likelihood of coating fracture rather than an indication of the true stresses in a system.

The effects of roughness and bond coat CTE will first be discussed to demonstrate general effects that are important to the detailed analysis of the effects of oxidation and creep that are presented later.

Roughness

Interfacial roughness has been the subject of study for TBCs [6, 8] and is a well known consideration for stresses in thin oxide films [14]. The case that is usually treated is for cooling of a two layer system that is assumed stress free at some high temperature, generally the maximum temperature of a thermal cycle. Petrus et al. [8] started with the assumption of a stress free condition at room temperature and then modeled one entire thermal cycle for a plasma sprayed TBC. They demonstrated that the assumption of a stress free

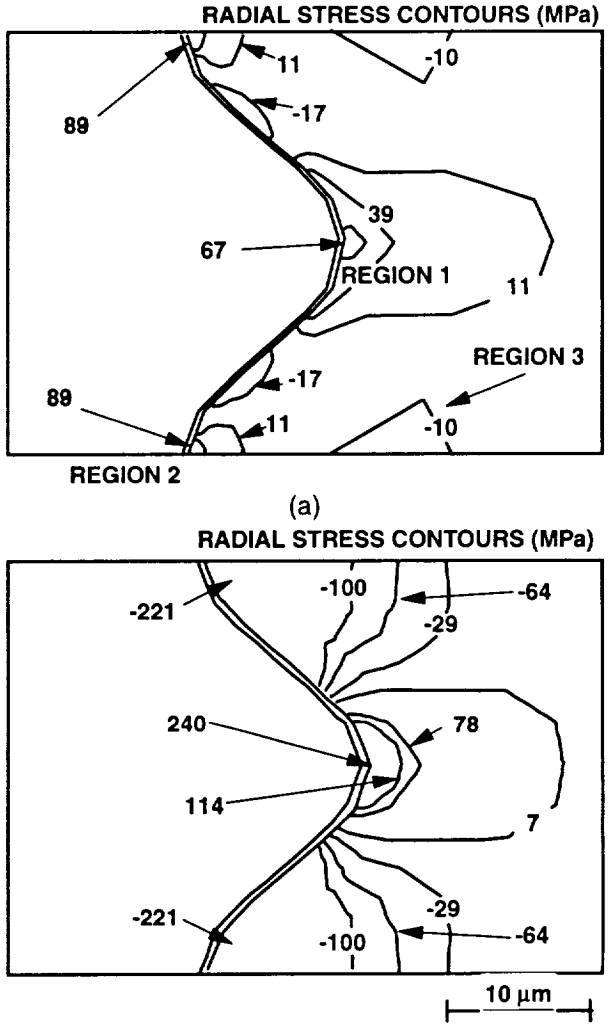


Figure 3. Contour plots of the radial stresses at a rough interface for (a) a low CTE bond coat and (b) a high CTE bond coat.

condition at high temperature is valid for a TBC if the bond coat and / or top coat relax sufficiently during a high temperature hold. When this holds true, Petrus et al. achieve qualitatively similar results as previous studies: a rough interface causes an increase in delamination stresses (tensile stresses normal to the interface) at the peaks of the metallic asperities while increasing compression normal to the surface in the valleys between asperities, as compared to a smooth surface. This finding was reexamined for the current study using four thermal cycles, instead of one, and including oxidation. It was found that, even when oxidation is considered, a smooth surface had stresses normal to the interface ranging from 5 MPa at the interface to 0.5 MPa at the surface. A rough surface, however, exhibited tensile delamination stresses as high as 67 MPa at bond coat peaks, and normal compressive stresses as high as 17 MPa in the valleys (Figure 3(a)). Tensile stresses were also developed in the valleys due to oxidation, as will be discussed later. Several points of interest arise from this finding: 1) roughness tends to cause a substantial increase in stresses in the near interface region as compared to a smooth interface, and 2) the stress is periodic and should tend to limit cracking to the bond coat peak regions. This result is qualitatively the same as for previous models that considered only cooling and only one thermal cycle. A third point is noted that the properties of the bond coat have nearly no influence on TBC stresses for the case of a smooth interface. However, bond coat properties become extremely important when the interface is rough [7, 8]. Note that roughness profiles, and hence the effect of roughness on stress, are extremely complex and at an early stage of investigation.

Bond Coat CTE

For the case of a smooth interface, it has been demonstrated that the properties of the bond coat are relatively unimportant. Rather, the substrate dominates stress generation in the coating. The strength of an effect in the presence of a rough bond coat is exemplified by the effect of bond coat CTE. Figure 3 shows the radial ceramic layer stresses for low CTE (Figure 3(a)) and high CTE (Figure 3(b)) bond coats and the standard conditions of oxidation and creep of the top coat and bond coat. The tensile stresses at the bond coat peak increase from 67 to 240 MPa with the increase in CTE from 14 x 10^{-6} $°C^{-1}$ to 20 x 10^{-6} $°C^{-1}$. Also, the valley stresses for the high CTE case are compressive, up to -221 MPa, while the valley stresses for the low CTE case tend to be mildly tensile. The factor of approximately three change in stress level at the peak scales well to the approximately factor of three change in CTE mismatch between the ceramic and the bond coat for these conditions. This large effect emphasizes the importance of roughness to the stresses in the near interface region.

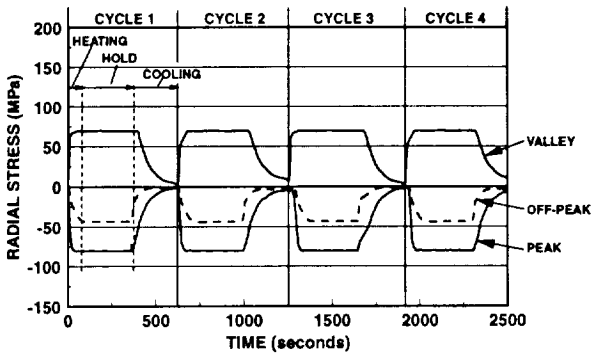
While it is tempting to suggest that a high CTE bond coat favors cracking at bond coat peaks and should therefore shorten TBC life, a high bond coat CTE also favors crack arrest over the valleys at this small number of cycles. As will be shown later, oxidation at larger numbers of cycles may have a dramatic effect on this situation. The combined effect of CTE and higher oxidation at large numbers of cycles has not yet been investigated, but it is known that CTE interacts with some of the other parameters studied [15].

The remainder of the discussion will focus on the generation of stresses due to the operation of oxidation and creep. The effects of the various mechanisms are best understood when presented separately, starting with the simple, all elastic case.

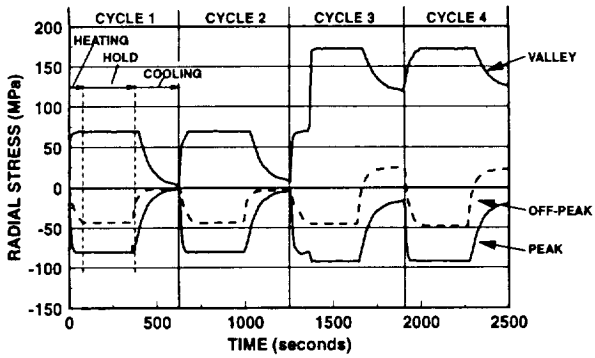
Elastic Case

In the case where all the constituents are elastic and there is no oxidation, the radial stress profiles for the three locations are the same for all four cycles (Figure 4 (a)). At the bond coat peak, the ceramic is driven into compression on heating and returns to zero stress on cooling. The stress in the off-peak location also cycles between a compressive stress and zero stress. However, the stress in the valley cycles between a zero cool-down stress and a tensile stress of approximately 70 MPa during steady state heating. The change in sign of stress from the peak to the valley is a result of the change in sign of the radius of curvature [14].

Oxidation is introduced incrementally in the model, with the first increment of oxidation starting with the start of the hold time in the third cycle. Oxidation changes the stresses significantly from the case of no-oxidation (compare cycles 1 and 2 to cycles 3 and 4 in Figure 4(b)). Oxidation pushes the peak radial stress toward slightly greater compression for all portions of the cycle and reduces the amplitude of the stress per cycle. Concurrently, the off peak cool-down stress becomes tensile while the off-peak steady state stress remains constant. This results in an increase in the cyclic stress amplitude for the off-peak location, where an increase in the stress amplitude may have an effect on the fatigue life of this material [16]. The most dramatic effect is on the stress in the valley. The valley stress increases rapidly with growth of the oxide. The linear, nearly vertical increase in stress reflects the linear growth rate imposed on the oxide. This rapid increase in stress is exaggerated due to the fact that all



(a)



(b)

Figure 4. Ceramic layer radial stresses at the peak, off-peak and valley locations for the case of (a) no oxidation and (b) oxidation starting at cycle 3.

of the oxide growth for the first three cycles is made to occur during a short time in the third cycle in the model. As is shown later, this exaggerated increase in stress is unimportant when creep is allowed to occur. The valley cool-down stress increased from zero to 120 MPa and the steady state stress increased from roughly 50 to 170 MPa, causing a substantial oxidation induced increase in the mean tensile stress. Figure 4(b) provides an indication of the stress levels that are expected if stresses due to oxide growth are not relieved. The change in stress amplitude for all locations is a result of the change in the system properties with the addition of a very thin oxide layer with a drastically different modulus and CTE than the coating materials.

This simple case illustrates a trend found throughout the results: oxidation tends to cause radial cool down stresses at the bond coat peak to become more negative, and off-peak and valley stresses to become more positive. Oxidation also causes a decrease in the peak and valley cyclic stress amplitudes and a concurrent increase in the off-peak stress amplitude.

The overall view in Figure 4(b) is that oxidation can have a significant effect on stress when no other mechanisms are active. Interestingly, the four cycle stress history in Figure 4 b would result in a system that could not support the commonly observed peak-to-peak cracking. This is due to the fact that the radial stresses are zero or compressive over the bond coat peaks for all 4 cycles. A real system, however, includes plasticity and creep (or, identically, stress relaxation) behavior that can modify these stresses substantially.

Creep Effects

Before examining the interactions of top coat and bond coat creep with oxidation, it is informative to revisit the effect of these mechanisms on stresses in the absence of oxidation [8] and to examine how these stresses evolve with thermal cycling. Figures 5 and 6 show the peak,

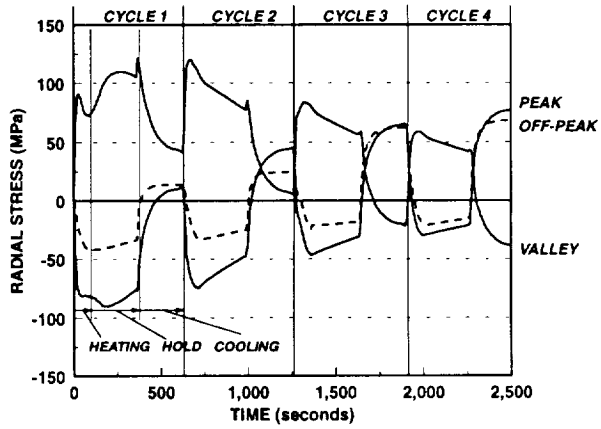


Figure 5. Variation in ceramic radial stress with position and time for the case of a creep in the ceramic and an elastic bond coat.

off-peak and valley stresses for systems with creeping top coat-elastic bond coat and creeping bond coat-elastic top coat, respectively. While the stress always returns to zero in the all-elastic case (without oxidation), creep of the top coat (Figure 5) causes a deviation from a zero cool-down stress at all locations. At the peak, the cool down stress progressively increases from the zero stress of the all-elastic case to roughly 70 MPa during the course of four thermal cycles. This gradually increasing cool-down stress is a result of partial relaxation of the top coat compressive stresses at the peak during each steady state hold (Figure 5). As with any relaxation process, the relaxation rate decreases with time (apparent as a decreasing slope during the hold time) and the cool-down stress tends toward a steady state value. However, relaxation is not complete by the end of the fourth cycle. The stresses at the off-peak and valley

locations also change as a function of time. The off-peak stresses become progressively more positive with cycling. The valley cool-down stress is actually more positive than the elastic case for the first cycle, but then becomes progressively more negative, eventually reversing sign by the fourth cycle.

The stress amplitude for each cool-down portion of the cycle remains constant with increasing cycling for each location, but the mean stress changes. The reason that the stress amplitude remains constant is that there has been no change in materials properties of the system during this process and relaxation is relatively inactive during the transients. The change in mean stress is due to the relaxation of stress during the steady state hold.

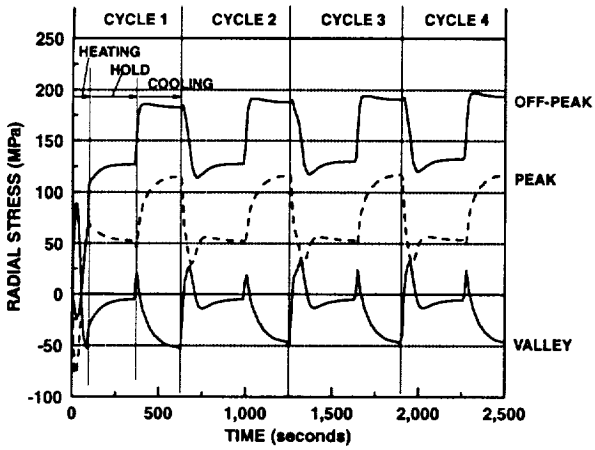


Figure 6. Variation in ceramic radials stress with position and time for the case of creep in the bond coat and an elastic top coat.

Bond coat creep has a similar effect to that of top coat creep, but the rate of the change is much higher simply because the bond coat creeps more readily (Figure 6) [2]. The magnitude of the change is also greater within the first four cycles. This is due to the fact that relaxation at steady state hold runs to completion for each hold cycle, including the first, resulting in stabilization of the thermal fatigue cycle after the very first cycle. Therefore, stress profiles for all three locations remain constant for the second through fourth cycles. This does not mean that bond coat creep stops after the first cycle. The evidence of continuing bond coat creep is apparent as a slight decrease in the stress during hold for all 4 cycles. Other work has also shown that bond coat creep is active for some materials during cool down on every cycle [2]. As with top coat creep, bond coat creep causes the peak and off-peak stresses to become more positive and the valley stress to become more negative.

Combined Effects

The combination of the effects of top coat creep, bond coat creep and oxidation are compared in Figures 7(a) - (c) as the radial cool-down stresses for the peak, off-peak and valley locations. First, note that the stresses

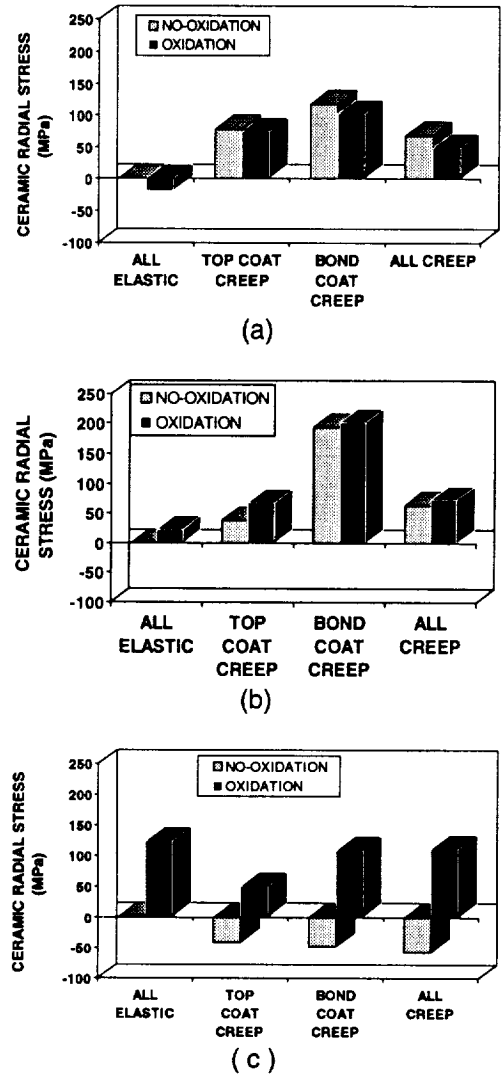


Figure 7. Ceramic radial cool-down stress variation for the cases of all-elastic, creeping top coat (elastic bond coat), creeping bond coat (elastic top coat) and "all creep" (combined top and bond coat creep) in oxidizing and non-oxidizing conditions for (a) the peak, (b) off-peak and (c) valley locations.

resulting from the combination of top coat creep and bond coat creep cannot be expressed by simple addition of stresses due to the mechanisms acting separately.

The peak stress is lower, the off-peak stress is intermediate and the valley stress is higher than would

be expected of either mechanism separately. This result illustrates the difficulty in looking at only one mechanism to characterize the stresses in a system that has more than one mechanism operating. The various creep cases do, however, remain consistent in the sign of the stress at each location.

Comparing oxidized to non-oxidized stresses shows there is interaction among mechanisms at the valley location but that oxidation and creep are roughly additive for the other locations. In the valley, oxidation reverses the stress from moderately negative in all three creep cases to highly positive. An interesting point is that the magnitude of the stress change is for the creeping bond coat and combined creep case is nearly double that for the ceramic creep case.

The results appear to indicate that the stresses at the peak and off-peak locations are tensile whether oxidation occurs or not. Thus, even if no oxidation occurs, the stress state at these locations is favorable to delamination cracking. The stress in the valley region without oxidation is compressive, clearly not conducive to cracking in this region, in agreement with previous considerations of wavy interfaces [14]. However, addition of oxidation changes the sign of the stress to become highly tensile, and conducive to delamination cracking, as noted by previous authors [6, 7]. It should be remembered, however, that the stresses reported in Figures 7(a)-(c) are from single elements quite close to

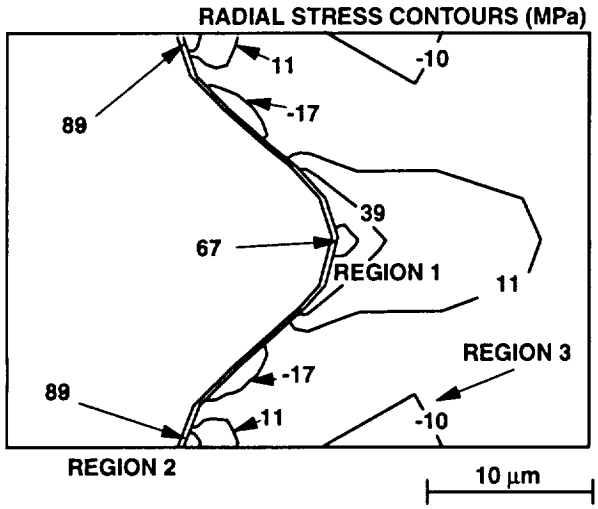
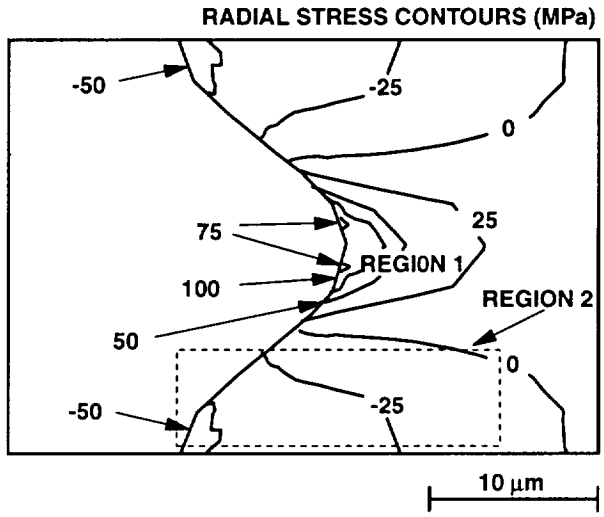


Figure 8. Contour plot of the radial stress in the ceramic after cool-down following 4 thermal cycles for the case of combined oxidation and creep.

the interface. Examination of a contour plot for the general creep case with oxidation, Figure 8 (same as Figure 3(a)), shows that the area of the peak, off-peak and especially the valley tensile stress regions are small and not connected. Also, a compressive region exists between the peaks (note Region 3 in Figure 8) that would prevent planar cracking from peak top to peak top, as is usually observed in failed coatings. Similarly, the high tensile stress regions are not in the same plane, so that cracks moving from one region to another would have to follow a path well off of parallel to the interface. Since cracks typically appear planar and parallel to the interface, this configuration does not agree with observations.



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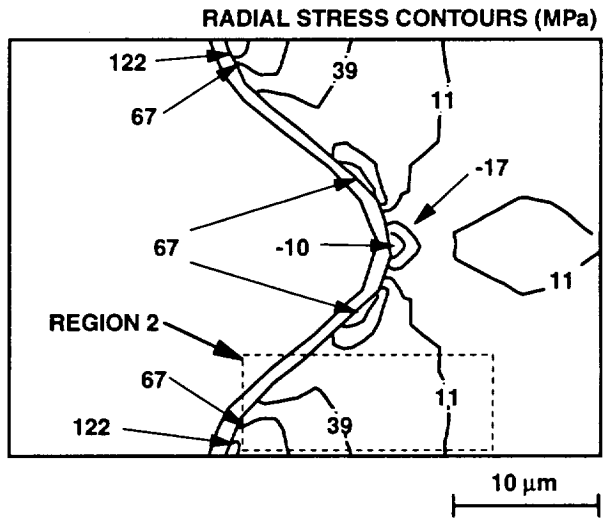

(b)

Figure 9. Contour plots of the radial ceramic stress at cool-down following 51 cycles for the case of combined creep and (a) without oxidation and (b) with oxidation.

This difficulty was resolved by running an additional combined creep case, with and without oxidation, to a greater number of thermal cycles. Figures 9 compares contour plots of the radial stresses after 51 thermal cycles for non-oxidizing and oxidizing cases. For 51 cycles without oxidation, a compressive stress region extends from the valley well into the ceramic layer (Region 2 in Figure 9 (a)). These compressive stresses would limit cracking to the region above the bond coat peaks. With oxidation, it is evident in both the 4 and 51-cycle cases (Figures 8 and 9(b), respectively) that the tensile stress at the peak has become compressive. In the 51 cycle case the tensile off-peak region has grown in area and become more tensile. Additionally, the compressive radial stress regions over the valley after 4 cycles with oxidation (Region 3 in Figure 8), and after 51 cycles without oxidation (Region 2 in Figure 9(a)), have completely disappeared after 51 cycles with oxidation (Figure 9b, Region 2). Thus, the stresses at 51 cycles with oxidation is much more favorable to crack growth over the valleys than is the four cycle case. However, the stresses at 51 cycles are unfavorable for crack growth at the peak of the bond coat. The histograms in Figure 10 provide a direct comparison of the stresses at each position for each case.

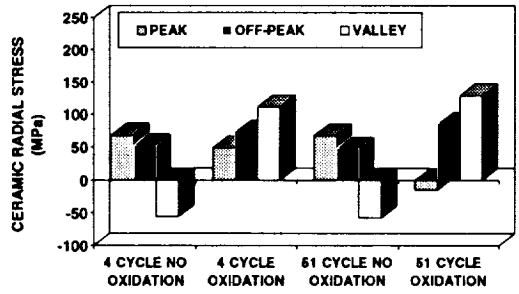

Figure 10. Radial stress in the ceramic at the end of cool-down as a function of location, number of thermal cycles and oxidation.

An overall view of the cracking process begins to emerge from these results. At small numbers of thermal cycles, the relatively rapid processes of ceramic and bond coat creep make cracking favorable at the peak and off-peak locations. However, cracking over the valley locations, except right at the interface, is still difficult due to compressive stresses and/or low tensile stresses. As the relatively slow process of oxidation proceeds, cracking over the peaks becomes less favorable while cracking at the off-peak and valley locations becomes easier. As oxidation proceeds even further, the regions above the valleys grow further until the tensile regions of neighboring valleys converge to form a uniform tensile region a short distance from the bond coat. Cracking in this large, general tensile region then becomes possible. Thus, it appears that early cracking should occur at bond coat peak regions, aided by ceramic and bond coat creep, and continued cracking is driven by the changes in stress over the valleys driven by oxidation.

Summary and Conclusions

The failure mechanisms of thermal barrier coatings have been examined through a FEM model of residual stress generation due to oxidation, top coat creep and bond coat creep. The results indicate that top coat and bond coat creep act to generate tensile stresses at bond coat peak and off-peak locations, while generating compressive stresses in the valley regions. The primary effect of creep on the cool down stress appears to be limited to short times at temperature. Oxidation acts opposite to creep, causing increasing tensile stresses in the valleys while pushing the peak regions into compression. Continuing oxidation results in an increase in the size of the tensile region associated with the valley, growing to the point that the tensile stresses in the valley extend beyond the peak regions. Continued oxidation also causes a slight increase in the maximum tensile stresses at the off-peak and valley locations.

The combination of oxidation, top coat creep and bond coat creep yields a stress distribution in time and in location along the interface that is not achievable by any one mechanism acting alone. The picture of the failure mechanism resulting from this evolution of residual stresses is as follows: 1) Early cracking at bond coat peaks, driven by tensile peak stresses generated due to creep processes. 2) Cracks generated early in life do not propagate due to creep generated compressive stress regions over the valleys. 3) At higher numbers of cycles, and therefore higher oxide thicknesses, stresses over the valleys become increasingly tensile and the size of the tensile region increases. 4) The tensile region over the valley is then capable of sustaining crack growth, resulting in linking of the near-peak cracks generated during early cycling. This description of failure is supported by the qualitative observations of cracking progression.

This progression of cracking is clearly a result of the combined action of creep, oxidation and thermal cycling. An accurate description of the entire process requires a model including these factors. However, failure of a TBC is very complex and is clearly not completely described by a simple model. Other factors, such as: sintering; phase changes in the oxide, bond

coat and ceramic layer; ceramic layer cracking; and compositional changes are also thought to be important aspects that should be included for a complete description of the failure mechanism.

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